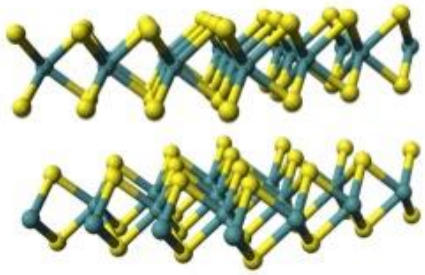
Center for the Computational Design of Functional Layered Materials (CCDM)
EFRC Director: John P. Perdew
Lead Institution: Temple University
Start Date: August 2014

Mission Statement*: To develop, apply and validate methods to calculate the electronic structure of layered and two-dimensional materials that have the potential for clean energy technologies.*

Research Plan*:* While these electronic structure methods can be useful for the design of many new materials, we focus on layered materials. We aim to predict how their properties are affected by composition, structure, interfaces, defects, and strain, and to design such materials to catalyze the evolution of hydrogen, a clean fuel, by solar-driven splitting of water. Validation and motivation are by experimental synthesis and characterization.

Background: New or modified materials with desired functionalities play an essential role in the development of clean-energy technologies, such as solar cells, batteries, and catalysts to generate hydrogen fuel by water-splitting. Because there are so many possible materials, it is impossible to grow and test them all in the laboratory. Computational design of materials with desired properties, based on first-principles theory and modeling, is a practical alternative. (The figure shows the structure of MX_2 layered materials).



Layered materials are typically composed of planar sheets, with strong bonds between the atoms within a layer and weak bonds between the layers. A collection of many layers is a three-dimensional (3D) material, while one or a few layers make a 2D material. Variation of properties with dimensionality is an appealing feature of these materials. The 2D materials can be grown on a substrate, or exfoliated from the 3D materials. Extraneous atoms can bind at the surface, or take up residence between the layers. The surfaces of many materials are contaminated by contact with air. The layered materials interact only weakly with the molecules in air, but can be modified by controlled exposure to free atoms.

Some layered materials have no fundamental energy gap. Graphene (unsupported 2D carbon) is a celebrated example. But interest for applications has shifted to “beyond-graphene” 2D semiconductors, such as boron nitride (BN) or molybdenum disulfide (MoS_2), and to heterostructures made by stacking different 2D crystals. Other transition-metal dichalcogenides (TMD’s MX_2, as pictured above), such as WS_2, WSe_2, and $MoSe_2$, are also of interest, as are layered oxides.

Layered 2D and 3D materials display many potentially useful properties. The direct energy gap in 2D MoS_2, which can be tuned by material modification, opens up the possibility of flexible optoelectronic or photovoltaic applications. Photo-excited electrons and holes can be spontaneously separated in a 2D heterostructure. A single layer of the 1T polytype of MoS_2 can catalyze the photo-induced splitting of water. Small ions can intercalate between the layers of a material, as in a lithium battery.

CCDM is organized into three thrusts and two cross-cutting fora:

- **Thrust 1. Theory.** *Density functional and correlated-wavefunction methods development*. A strong component of fundamental methods development and testing is a unique contribution of our Center.
- **Thrust 2. Computation and Modeling.** A. *First-principles computation and design of layered materials*. B. *Multi-scale modeling with first-principles input for extended defects in layered materials*.
- **Thrust 3. Synthesis and Characterization.** A. *Growth and characterization of layered materials*. B. *Catalysis and water splitting in layered materials*.
- **Cross-cutting Forum A.** *Properties of layered and 2D materials.*
- **Cross-cutting Forum B.** *Applications of layered and 2D materials.*

Some major achievements of the first two years: Accurate but efficient density functionals including van der Waals interaction have been developed and extensively tested. Also higher-level methods for strong correlation, including robust coupled-cluster and particle-particle random phase approximation.

Bending a 2D material has been computationally predicted to have major effects on electronic structure and thus on the electrical conductivity and catalytic properties. Some 2D materials have been predicted to be topological insulators. Multi-scale and continuum models have been developed to describe extended defects, growth and phase transformations.

Chemical vapor deposition techniques have been developed to grow uniform MoS_2 films or highly-ordered islands. A charge-density wave has been observed in few-layer $TiSe_2$ by TEM. The composition and crystallinity of MoS_2 can be engineered for catalytic performance. Joint computational/experimental studies have shown that intercalation of Cu atoms and Ni ions can transform the inexpensive layered material birnessite from an inefficient water oxidation catalyst into a very active one. A new catalyst for the oxidation step of water-splitting, $CoTe_2$, has been prepared.

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